Radiative Lifetime of Metastable Li II 2^1S_0

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The radiative lifetime of Li II 2^1S_0 has been measured by counting decay photons versus time from an ensemble of metastable Li ions stored in a simple electromagnetic trap of the Penning variety. The result is $\tau_{expt} = 503(26)$ μsec. The error represents one standard deviation from the mean of a series of 34 separate determinations. This result is in agreement with the theoretical value $\tau_{th} = 513$ μsec.

The 2^1S_0 state of Li II lies 60.7 eV above the 1^1S_0 ground state and, as in all He-like systems, may decay to the ground state primarily by emission of two electric dipole photons. The rate[1] for such a process is about $\alpha(Z\alpha)^2$ ($\approx 10^{-6}$ for Li II) times a single-photon electric dipole rate, which accounts for the metastability of 2^1S_0 in He-like species. [The forbidden magnetic-dipole decay rate to the nearby 2^3S_1 is extremely small ($\sim 10^{-8}$ sec^{-1} in Li II) and makes no significant contribution to the 2^1S_0 lifetime.] The two-photon decay rate for the 2^1S_0 state of systems from He I to Ne IX has been calculated by Drake, Victor, and Dalgarno.[2] They obtain for Li II $\tau_{th} = 513$ μsec. Previous experimental measurements using different methods have reported results for He I[3,4] and Ar XVII.[5] The two values for He I differ widely, being (from Ref. 3) 38(8) msec and (from Ref. 4) 19.7(10) msec. There is good agreement between the theoretical predictions, the results of Ref. 4, and the Ar XVII measurement. However, because shielding of the nuclear charge plays a less significant role in the theoretical calculation of τ with increasing Z, it is conceivable that the Ar XVII measurement could be consistent with the result of Ref. 3 provided there were a suitable adjustment of the theoretical calculation.

This Letter describes work utilizing an ion-storage method to determine τ for Li II. The result is in agreement with the calculations of Drake, Victor, and Dalgarno[2] and thus suggests resolution of the discrepancy between the two He I measurements in favor of Ref. 4. To the authors' knowledge, this is the first application of the ion-storage technique to the measurement of a metastable lifetime.

The method used is manifestly straightforward. A quantity ($\approx 10^2$) of Li ions in the 2^1S_0 state is created at $t=0$. They are stored in an ion trap in a region (≈ 3 cm^3) viewed by photon detectors capable of counting a constant fraction of the decay photons which leave the region during the confinement time. These counts are accumulated over several mean lifetimes after $t=0$. All ions are then swept from the trap, and a new cycle is begun. Many fill-store-dump cycles are repeated until a decay curve is built up which has sufficiently small statistical error to allow determination of a mean lifetime. This is the scheme in its barest essentials; the details follow.

Figure 1 shows the ion-storage trap and the photon detectors. Except for the electromagnet, the entire apparatus is maintained under vacuum with a base pressure of $\lesssim 1\times10^{-8}$ Torr. During operation the pressure may rise to 1×10^{-7} Torr, and is sometimes purposely allowed to rise as high as 1×10^{-6} Torr. The vacuum envelope is primarily stainless steel. The ion-storage volume is located in the center of the axially symmetric electrode structure between electrodes 2 and 3. A large magnetic field (3.8–8.5 kG) confines the ion motion to be near the axis of the magnetic field, and negative potentials on electrodes 2 and 3 with respect to 1 and 4 confine the

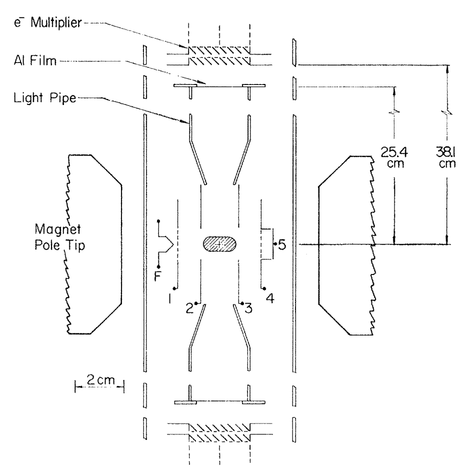

FIG. 1. Sketch of the ion trap and photon detectors. The neutral Li beam is represented by the cross-hatched area in the center and is directed out of the plane.

ions to the region between 2 and 3.

Li ions are created inside the trap by electron bombardment of a Li atomic beam. The beam is depicted in Fig. 1 by the cross-hatched region in the center and is directed out of the plane of the figure. The neutral Li beam intensity is monitored by a surface ionization detector mounted above the plane of Fig. 1. Electrons are emitted by a directly heated thoriated tungsten filament F, and accelerated along the magnetic field by a negative potential applied to F.

During any portion of the duty cycle the electrical configuration of the trap can be specified by the potentials applied to each of five electrodes as the list $(V_F, V_1, V_2, V_3, V_4)$. Electrode 5 does not appear in the list since it is always at the same potential as 4 and serves only as a collecting electrode for electrons or dumped ions. A typical duty cycle consists of the following sequence: a fill portion lasting 22 μsec during which the list is $(-160, 0, -12, -12, 0)$, a storage period of 2 msec with the configuration $(+20, 0, -12, -12, 0)$, and a dump period of 60 μsec with $(+20, +6, +6, 0, 0)$. These values are typical of those most used to obtain decay data; however, in searching for systematic effects they were varied widely.

Conservation of energy requires that the two photons emitted in the 2^1S_0 decay have energies such that $h\nu_1 + h\nu_2 = E(2^1S_0) = 60.7$ eV. Thus the distribution in wavelength of single photons issuing from an ensemble of 2^1S_0 ions is a continuous one with a short-wavelength cutoff at $\lambda_{min} = 204$ Å and extending to infinite wavelength. For Li II, this distribution[2] shows a sharp rise from λ_{min} to a peak at $\lambda = 258$ Å, after which the intensity drops off roughly as $\exp[-(\lambda - 258)/200]$.

To detect this radiation, two EMI 9642/2 eighteen-stage CuBe venetian-blind electron multipliers are used as shown in Fig. 1. To prevent metastable neutral molecules from reaching the multipliers, their view of the storage volume is covered by aluminum films 0.75 in. in diameter and 800 Å thick. These films have "good" transmission $(10\% < T < 70\%)$ over the range 200 to 700 Å. It is estimated that the Al film–CuBe multiplier combination responds to ~2% of the radiation over the region 200 to 500 Å which strikes the Al film. The multipliers are protected from the stray field of the electromagnet by shielding and by placing them ≈38 cm from the center of the magnet gap. To regain some of the solid angle lost by such a large separation from the trap center, two "light pipes" were used. They are made of Pyrex and coated internally with a 1000-Å thickness of gold.

Pulses from the multipliers are gated into the memory of a multichannel scaler during the trap storage period. The memory is stepped through 100 channels with 20 μsec dwell per channel. Completion of the last channel signals the end of one storage-and-data-collecting period. In order to compensate for counts which originate from excitation of background gas ions and neutrals, a movable beam stop is alternately moved into and out of the Li atomic beam. The stop is held in each position for 2000 passes through the 100-channel memory block (≈4 sec total). Counts collected with the beam on and off are stored in separate 100-channel blocks. The time base for the system is derived from a 100-kHz crystal oscillator.

The trap, operating with background pressures of $\sim 5\times10^{-8}$ Torr, will easily store $\approx 10^6$ ions with a mean decay time of ≈1 sec. More sophisticated traps[6] have achieved much longer storage times, but the apparatus described here is quite adequate for the job at hand, and has the advantage of simplicity and an open structure to allow exit of the decay photons. It should be pointed out that, even under optimum conditions, the number of Li ions stored was never more than about 10% of the total number of stored positive ions. The majority of ions (determined by cyclotron resonance to be principally N_2^+) were created from the background gas. Although overwhelming in number, their effects were easily separated from those due to Li ions by means of the beam stop and their presence did not prejudice the results.

Figure 2 shows a decay curve representative of the 34 used to determine τ. During collection of these data, the trap potentials were as described in the example above and the magnetic field was 7.3 kG. The curve is the result of computing the channel-by-channel difference of the Li beam-on and Li beam-off data. The ratio of counts with beam on to beam off was ≈7:1; the collecting time was 17 min which, taking into account the ≈25% duty cycle, corresponds to an average detected decay rate of 32 counts/sec. This rate implies that the number of 2^1S_0 ions stored during any one cycle averaged about 300. This number is only an estimate and might vary up or down by a factor of 10 depending on the largely unknown detector-efficiency and solid-angle factors.

Threshold studies of the decay rate versus

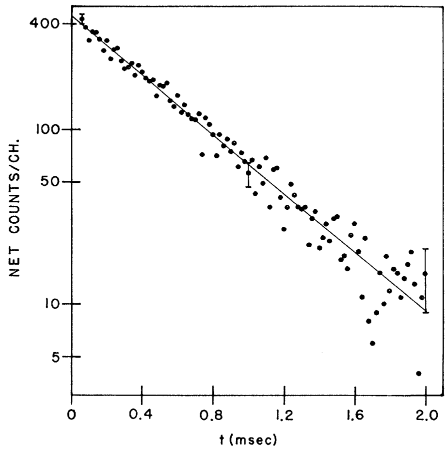

FIG. 2. A representative Li II 2^1S_0 decay curve. The line through the points is the computer fit which gave $\tau = 511$ μsec. The final result is a mean value derived from 34 such runs.

electron impact energy showed an onset at 66(4) eV in agreement with the threshold of 66.1 eV for Li I $+e^- \rightarrow$ Li II $(2^1S_0)+2e^-$. Of course, 2^3S_1 ions (threshold 64.4 eV) are also created and stored in roughly the same numbers as 2^1S_0. However, their decay rate by forbidden $M1$ is $\approx 10^{-8}$ times the 2^1S_0 rate. Hydrogenlike metastable Li III $(2^2S_{1/2})$ should not be present in any significant amount for impact energies below 172.8 eV and not at all below 91.8 eV. Thus, it is reasonable to conclude that the decay observed is that from 2^1S_0.

The data were fitted with the function $Ae^{-t/\tau}+B$ by a least-squares computer routine which yielded the parameters A, B, and τ. The origin of the base line B is not certain at this time, but it is never more than $\simeq 0.01A$. Although it is a small fraction of A, its reality has been established by runs made with storage time extended to 16 msec. B may arise from quenching collisions of 2^3S_1 ions with background gas atoms. Further work is planned to understand this phenomenon.

In a search for systematic effects, the following quantities were varied over the ranges indicated: trapping voltage $(V_{2,3})$ from -6 to -18 V, magnetic field from 3.8 to 8.5 kG, electron impact energy from 88 to 268 eV, and Li beam intensity from $\approx 5\times 10^{10}$ to $\approx 5\times 10^{12}$ cm^{-2} sec^{-1}. No significant variations in the fitted τ's were discovered. In addition, the background gas pressure was increased from $\approx 5\times 10^{-8}$ to $\approx 1\times 10^{-6}$ Torr with a resultant 10% reduction in the fitted lifetime. This is considered good evidence that, at the lower pressure where final data were taken, collisional shortening of the 2^1S_0 lifetime was not important.

TABLE I. Current status of measured 2^1S_0 lifetimes.

System	Experiment (sec)	Ref.	Theory (sec)	Ref.
He I	$3.8(8)\times 10^{-2}$	3	1.95×10^{-2}	2
	$1.97(10)\times 10^{-2}$	4		
Li I	$5.03(26)\times 10^{-4}$	This work	5.13×10^{-4}	2
Ar XVII	$2.3(3)\times 10^{-9}$	5	2.34×10^{-9} [a]	[5]

[a]From the asymptotic (large-Z) relation $1/\tau = 16.46\times(Z-0.797)^6$ sec^{-1}, due to G. W. F. Drake in a private communication to Marrus and Schmieder (see Ref. 5).

It was discovered that the fitted τ varies inversely with the number of stored charges once this number exceeds some maximum amount. This amount in turn varies with the trap parameters. For the set of parameters used to obtain the final data reported here (see discussion of Fig. 2), this number was $\approx 6\times 10^6$ positive charges. The effect presumably arises from ion-ion collisions which either quench the 2^1S_0 state to the ground or 2^3S_1 states, or cause the Li ions to gain enough energy to leave the trap. It may also result from distortion of the trapping fields due to the ion space charge. In any case, when the ion density is kept below this critical value, no effect on τ is observed and all the final data were taken in this safe area.

The result for $\tau(2^1S_0)$ is 503(26) μsec. This is the mean of 34 separate determinations and the error is 1 standard deviation. Table I summarizes the current status of measured 2^1S_0 lifetimes. It is seen that, except for the results of Ref. 3, the agreement between experiment and theory is good, extending over a range of 10^6 in lifetime.

Further work is continuing to improve the number quoted here and to exploit the ion-storage technique for the study of metastable lifetimes in other positive and negative ions.

[1]H. A. Bethe and E. E. Salpeter, *Quantum Mechanics of One- and Two-Electron Atoms* (Academic, New York, 1957), Chap. 4, p. 285.

[2]G. W. F. Drake, G. A. Victor, and A. Dalgarno, Phys. Rev. 180, 25 (1969).

[3]A. S. Pearl, Phys. Rev. Lett. 24, 703 (1970).

[4]R. S. Van Dyck, Jr., C. E. Johnson, and H. A. Shu-

gart, Phys. Rev. Lett. 25, 1403 (1970), and Phys. Rev. A (to be published).

[5]R. Marrus and R. W. Schmieder, to be published.

[6]H. G. Dehmelt, in *Advances in Atomic and Molecular Physics*, edited by D. R. Bates and I. Estermann (Academic, New York, 1967), Vol. III, pp. 53-72

Observations of Moving Self-Foci in Sapphire*

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Observations of the time development of damage tracks in sapphire are explained in terms of a backward-moving self-focused spot. The detailed characteristics of the dynamics of the track evolution depend on the temporal shape of the incident laser pulse. Both qualitative and semiquantitative features of the observed phenomena are explained using a theory which shows that the results are consistent with a combination of both electrostrictive and electronic self-focusing mechanisms.

The importance of self-focusing in enhancing optical intensities in solids to material-rending levels has long been recognized.[1,2] Unfortunately, it has been difficult to study the self-focusing process quantitatively in solid materials because, at the very high incident power levels required, the source-beam quality is ofter poor and not reproducible. In this note, we present experimental data on the space and time evolution of damage tracks in sapphire, induced with a high-quality, reproducible, ruby laser-amplifier source.[3] We show that these tracks may be understood qualitatively, and to some extent quantitatively, as forming at the moving focus of the self-focusing source beam.[4] Experiments supporting the existence of a moving self-focus in nonlinear liquids have been reported previously.[5] It is hoped that this unambiguous evidence of a moving self-focus in sapphire will help clarify the interpretation of other "filamentary" phenomena observed concomitantly with self-focusing in other media.

The experimental arrangement has been described in detail elsewhere.[3] All experiments were performed using as a source a mode-controlled, Q-switched ruby oscillator and amplifier. The far-field beam profile was measured to be Gaussian down to 8% of the peak using a modified multiple-lens camera technique.[6] The light was focused into the sample using a lens ($f = 19$ cm) designed for minimum spherical aberration. The sapphire samples were typically 3-in.-long by 0.25-in.-square bars. Fast streak photographs were taken using an STL image-converter camera operating in the streaking mode. In most experiments a Corning 4-94 filter was placed between the camera and the sample which blocked light at 6943 Å, passing only the blue-green portion of the broad-band light from the self-luminous damage track. In addition a portion of the main beam was allowed to enter the camera directly, giving a marker streak relating the time of formation of a particular point on the damage track to the peak of the incident pulse.

Figure 1(a) shows a typical damage track. It has a "head" of relatively massive damage followed by a tapering tail which always ends at or

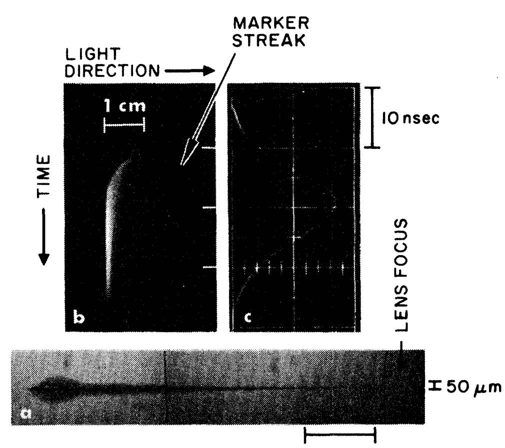

FIG. 1. Typical examples of (a) damage filament, (b) streak photograph, and (c) oscilloscope trace for a temporally smooth incident pulse.